



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In the Application of: Matthew Levinson )

Serial No. 10/680,541 )

Filed: September 7, 2003 )

For: ARTICLES AND METHODS FOR  
TREATING FABRICS BASED ON  
ACYLOXYALKYL  
QUATERNARY AMMONIUM  
COMPOSITIONS )

Examiner: John R. Hardee )

Art Unit: 1751 )

CERTIFICATE OF MAILING

I hereby certify that this correspondence is being deposited with the United States Postal Service as express mail in an envelope addressed to: Mail Stop Amendment, Commissioner of Patents, P.O. Box 1450, Alexandria VA, 22313-1450 on:

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37 C.F.R. §1.132 DECLARATION OF DR. MATTHEW LEVINSON

Dear Sir:

I, Matthew Levinson, of Lincolnwood, Illinois 60646, declare as follows:

1. I have the following education and experience:

B.A. Chemistry, Rutgers University, New Brunswick, NJ June 1978;

Ph.D. Chemistry, University of Pennsylvania, Philadelphia, PA December 1984;

Research Chemist, FMC Corp., Princeton, NJ 8/1983 – 5/1986;

Sr. Research Chemist, ANGUS Chemical, Buffalo Grove, IL 6/1986 – 6/1987;

Sr. Research Chemist, Stepan Company, Northfield, IL 6/1987 – 3/1992;

Research Fellow, Stepan Company, Northfield, IL 3/1996 – 12/1999;

Director of Research & Development for Consumer Surfactants, Stepan Company, Northfield, IL  
1/2000 – 6/2002; and

Vice President, Product Development – Surfactants, Stepan Company, Northfield, IL since June,  
2002.

3. I am named as an inventor on the patent application identified above.

4. I have reviewed the above-identified patent application and the Office Action mailed by the Patent Office on March 31, 2004. In making this declaration I have specifically reviewed DE OS 3,818,013 A1 ("the Voelkel reference"), and claims 1, 3-5, 12-15, and 17-19 of the above-identified patent application.

5. Additionally, I attended an interview with Examiner Hardee on March 4, 2004, regarding the instant application and the "Voelkel" and "Corona" (U.S. Patent No. 5,503,756) references. During the interview, it is my understanding that the Examiner removed the rejection of the pending claims in light of the Corona reference, but maintained the rejection of those same claims in light of the Voelkel reference. The Examiner, however, suggested that he would reconsider the rejection under Voelkel in light of proof of unexpected results over the exact formulation disclosed in Example 1 of Voelkel.

6. I refer to and incorporate by reference my 37 C.F.R. §1.132 declaration of March 13, 2000, which was filed in connection with the prosecution of U.S. Patent Application Serial No. 09/397,237 before the U.S. Patent Office (now abandoned), a copy of which is attached hereto as Exhibit A.

7. The present declaration differs from my previous declaration in that a different cationic fabric softening compound was used in reproducing the composition of Example 1 of the Voelkel reference, as described in detail below, and as requested by the Examiner during the

interview of March 4, 2004.

8. In summary, the claims of the above-identified patent application are concerned with dryer sheets and more specifically with dryer sheets that contain acyloxylalkyl quaternary ammonium compositions that comprise glycerol and esters thereof, in particular glycerol monostearate ("GMS") and esterquats. As shown by the results presented below, these products quite unexpectedly do not suffer from a condition referred to in the dryer sheet industry as "box bleed."

9. "Box bleed" is the melting or transfer of the fabric softener or conditioning material embedded in the dryer sheet substrate onto the box in which the fabric softening dryer sheets are packaged and sold to the consumer. This condition can occur when boxed dryer sheet products are exposed to temperature conditions that exceed the softening point of the active mixture coated on the substrate. The result of this softening can first be manifested by the migration of active ingredient from higher sheets to lower sheets, depleting the actives from the substrate in the upper portion of the box, and enriching the substrate in the lower portion of the box relative to vertical orientation.

10. Migration of the active ingredient can result in non-uniform performance during consumer use of the product. Further, migration of the active ingredient can result in adhesion of the adjacent sheets, making it difficult to dispense the unit substrate from the box or to peel a sheet off of a roll. In some instances, the active ingredient wets the cardboard packaging, causing staining and discoloration on the interior and exterior of the box, and potentially causing staining to adjacent boxes in the case in which the box is packed for transport to the retailer.

11. It is my understanding and belief that box bleed causes a consumer perception of a defective product, resulting in lost sales at the retailer and additional shipping and handling

costs associated with removal and disposal of the damaged package. For example, commercial products such as Bounce® and Snuggle® have not been able to fully overcome the box bleed drawback, and some manufacturers have been forced to apply a barrier coating to the inside surface of the cardboard box to prevent staining, resulting in higher packaging costs. Furthermore, box bleed is a drawback that incurs real costs for the manufacturer and retailer of dryer sheet products.

12. As a result, I understand that box bleed is an extremely undesirable characteristic from a consumer point of view, and it has been a consistent drawback of dryer sheet fabric softening compositions.

13. Applicant's invention surprisingly and substantially reduces or prevents active fabric softener ingredient "bleeding" from the product substrate when packaged as a final product within a cardboard or other paper-based "box", even during extended storage at elevated warehouse or trucking temperatures (See Specification, page 4, lines, 11-12; Table 2, *infra*).

14. In contrast with the claimed compositions, the examples described in the Voelkel reference produce excessive box bleed.

15. The following tests were conducted under my supervision and/or control to demonstrate the nonobvious properties of the claimed compositions.

16. Preparation of Voelkel composition and coating mix

A composition meeting all of the requirements identified by the Voelkel reference was prepared under my direction with the following characteristics: (i) The biodegradable fabric softener (a1) prepared, dimethyl di(oleoylisopropyl)ammonium methosulfate, is the identical fabric softener compound defined by the Voelkel reference, Example 1. (This cationic compound differs from the cationic compound prepared in my §1.132 declaration previously

filed in U.S. Patent Application Serial No. 09/397,237 ("the '237 Application"). The previously prepared cationic compound prepared pursuant to my declaration in the '237 Application was dimethyl di(oleoylethyl)ammonium methosulfate.). (ii) The biodegradable fabric softener (a1) was prepared from fatty acid groups that fall within the composition range stipulated by the Voelkel reference. (iii) The fabric softening ingredient (a1) prepared by the Applicant was fluid at room temperature as stipulated by the Voelkel reference. (iv) The fabric softening ingredient (a1) had a softening point in the defined range of 35-60°C when blended using the formulation provided in Example 1.

The reaction parameters, antioxidants, catalyst, and solvent used in the preparation of the esteramine intermediate and the resultant cationic compound are all well known in the art. The quaternization reaction of the esteramine intermediate was carried out in the absence of solvent which resulted in a cationic compound that is fluid at room temperature. Stearic acid was then added to the cationic compound to improve its handling properties. This solvent system was taken into account when preparing the rest of the example as described below.

17. Preparation of Dimethyl di(Oleoylisopropyl) Ammonium Methosulfate

To a 4-neck reaction flask equipped with a thermocouple, stirrer, nitrogen sparge line, a distillation apparatus with receiver, and a dry ice/acetone trap was charged oleic fatty acid (640 grams, 2.2706 mol comprising approximately 70% oleic, 11% linoleic, 7% palmitoleic, 4% palmitic, and 5% minors). A heating mantle was attached, and the vessel contents were warmed to between 60-70 C. To the flask was charged N-methyl diisopropanolamine (MDIPA, 180 grams, 1.2272 mol), and Anox® 20 (0.82 grams), which is an antioxidant commercially available from Great Lakes Chemical Company, Evansville, IN. The resultant mixture was stirred and heated to a temperature of approximately 100°C under nitrogen flow, when

0.81 grams of hypophosphorous acid (50% solution in water) was charged to the reaction flask. The resultant mixture was stirred and heated to a temperature ranging between 170-175°C under a nitrogen sparge for about 23 hours, and water resulting from the condensation reaction was collected through the distillation apparatus in the receiver. After 23 hours, further water removal was achieved by first draining the receiver, then applying vacuum to achieve a pressure of approximately 50 mm Hg in the reactor for about 1.5 hours, at which time the vacuum was released with nitrogen and the reactor contents were cooled to a temperature between 50-60°C. Analysis of the free fatty acid content confirmed that the esterification reaction had reached a typical degree of completion, with an unreacted fatty acid level of 0.0365 meq/gram (approximately 1% w/w based on a MW=275 amu). The intermediate thus obtained, methyl di(oleoylisopropyl) amine (hereafter referred to as "esteramine", 781 grams, 1.205 mol) exhibited a Gardner color of about 4, and was analyzed by perchloric acid titration (PAT) to confirm its molecular weight and to calculate the theoretical quantity of dimethyl sulfate (DMS) required to convert >95% of the esteramine to the quaternary salt.

The quaternization reaction was conducted by charging a portion of the esteramine (651 grams, 1.004 mol) to a stirred reaction vessel, heating the portion of the esteramine to a temperature of approximately 60°C, and slowly adding dimethyl sulfate (DMS, 122 grams, 0.968 mol), while applying cooling as required so that the reaction temperature did not exceed 90°C over the course of about 1 hour. Upon completion of the DMS addition, the contents of the flask were allowed to react for an additional 5 hours at a temperature of between 85-90°C. Complete consumption of the DMS was demonstrated by a negative DMS Draeger tube test of the vapor space above the reaction mixture. The resultant product, dimethyl di(oleoylisopropyl)ammonium methosulfate (773 grams, 0.999 mol) was maintained at 85°C,

while a 94 gram retain sample was removed from the reactor. The balance of the material in the reactor, approximately 678 grams (0.876 mol), was diluted with stearic acid, (291 grams), to improve viscosity at handling temperature. The resultant mixture comprised approximately 70% dimethyl di(oleoylisopropyl)ammonium methosulfate diluted with approximately 30% free fatty acid. The mixture was analyzed for unreacted free amine content via PAT titration and demonstrated a low level of 0.0420 meq/gram (approximately 3.28% w/w based on a MW=780 amu), confirming reaction with DMS.

#### 18. Preparation of Voelkel Coating Mix

All of the ingredients that were solid at room temperature were melted at their respective melt points prior to their incorporation into the coating mix. Approximately 12.85 grams of additional hardened tallow fatty acid was added to approximately 57.15 grams of the above described mixture comprising approximately 40.00 grams of dimethyl di(oleoylisopropyl)ammonium methosulfate and approximately 17.15 grams of free fatty acid to bring the free fatty acid to the proportions specified by Voelkel in Example 1. The resultant mixture was agitated until homogeneous. Then, approximately 20.00 grams of glycerol monostearate ("GMS") and approximately 5.00 grams of ethylene glycol stearate were added to the mixture and blended until homogeneous.

Next, approximately 0.50 grams of citric acid was added to the blend and mixed until homogeneous. All mixing steps were carried out at a temperature of approximately 80°C. Then, approximately 4.50 grams of perfume was added to complete the coating mixture of Example 1 of the Voelkel reference prior to application to the substrate as described below. The components of Example 1 of the Voelkel reference are shown as Composition A in Table 1. Composition B, which was prepared for this declaration, is identical to Composition A, but was assembled as

described above to minimize the viscosity of the liquid softener and ensure thorough mixing of all components at the lowest possible temperature.

**Table 1**

	<b>A</b>	<b>B</b>
<b>Constituent</b>	<b>Voelkel Example 1, wt. %</b>	<b>Comparative Example Prepared as set forth above Coating Mix<sup>1</sup>, wt. %</b>
Liquid Softener (a1)	40.00	40.00
Additional Hardened Tallow Fatty Acid	30.00	12.85 <sup>2</sup>
Hardened Tallow Fatty Acid from (a1)	N/A	17.15
Glycerol Monostearate (GMS)	20.00	20.00
Ethylene Glycol Stearate	5.00	5.00
Citric Acid	0.50	0.50
Perfume	4.50	4.50
Total	100.00	100.00

**19. Preparation of Composition of Example 5 of the Above-Identified Patent Application**

The method of preparation and box bleed testing results for the composition set forth in Example 5 of the above-identified patent application are incorporated by reference from my previously filed declaration submitted during the prosecution of U.S. Patent Application Serial No. 09/397,237, a copy of which is attached as Exhibit A. The method of preparation and box bleed test results of my previous declaration utilized an identical Example 5 formulation and are used for comparative reference in the instant declaration and prosecution of the above-identified patent application.

Briefly summarized, the three compositions prepared according to Example 5 of the above-identified application had the following compositions.

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<sup>1</sup> This composition is identical to Voelkel Example 1. Since the Liquid Softener (a1) used in B already contained some Hardened Tallow fatty acid, only 12.85 grams of additional Hardened Tallow Fatty Acid was necessary to make up the total of 30 grams in the complete mixture.

<sup>2</sup> Total Hardened Tallow Fatty Acid is 30.00% of the complete mixture.

	Invention Composition 1, wt. %	Invention Composition 2, wt. %	Invention Composition 3, wt. %
Triethanolamine-Hard Tallow-Based Ester Quat ("TEAHTEQ") of Example 5 Mixture	72.19	62.79	53.44
GMS present in Example 5 Mixture	24.06	20.96	17.81
Additional GMS	0	12.50	25.00
Free perfume	3.75	3.75	3.75
Total	100.0	100.0	100.0

## 20. Preparation of Fabric Conditioning Sheets

Fabric softening dryer sheets were prepared by impregnating a flexible non-woven substrate sheet with a liquefied organic fabric conditioning mixture. Individual impregnated articles were approximately 7 x 5.25 inches in size containing 1.25 +/- 0.5 grams of the impregnated mixture. The flexible substrate was impregnated by depositing the liquefied fabric conditioning mixture on a metal plate covered with foil, which was heated on a hot plate to the melting temperature of the mixture. The mixture was spread on the foil by a metal rod heated to the metal plate temperature. The substrate was placed on top of the mixture, covered by another layer of foil and pressed with a hot clothing iron to ensure uniform distribution of the fabric conditioning mixture over the substrate. The substrate was flipped to impregnate both sides of the sheet. The resulting dryer sheets were cooled and allowed to stand at room temperature overnight.

## 21. Box Bleed Test

A modified version of the fabric staining "Sandwich" Test described in U.S. Patent No. 4,073,996 (See column 16) was employed to determine the extent of box bleed of each product. In this test, simulated storage conditions were created to evaluate the degree of bleeding

provided by each of the compositions tested. The test involved building a "sandwich" with the fabric softening dryer sheet article at the center and heating this sandwich to elevated storage temperatures for an extended period of time.

The sandwich was made using a flattened typical dryer sheet box (approximately 7 x 7.25 inches), which was weighed before the start of the experiment. External to the box were Bakers Secret Teflon-coated cookie sheets (to prevent the escape of the bleeding product). Within the box ("the center") was a stack of ten flexible substrate dryer sheet articles (approximately 7 x 5.25 inches) prepared according to procedure described above. This "sandwich" was pressed together with a three pound weight and was placed in a constant temperature oven at approximately 50°C for one week (seven days). The "sandwich" was then removed and cooled. The center was removed and the box re-weighed. Any increase in weight corresponded to the approximate amount of the fabric softener composition that bled onto the box. The results of this test using the Comparative Examples B and D are set out in Table 2, below.

**TABLE 2**

BOX BLEED RESULTS 50°C Oven	Blank <sup>1</sup> (average of 2)	Comparative Example B (Voelkel, Example 1)	Composition 1	Composition 2	Composition 3
One Day (visual)	-----	Some bleeding	NO bleeding	NO bleeding	NO bleeding
One Week (visual)	-----	Profuse bleeding	NO bleeding	NO bleeding	NO bleeding
Original Box Weight	26.60	26.54	26.97	26.74	26.49

<sup>1</sup> Blank is a flattened typical box as described above with no substrate article. The change in box weight that is observed in the Blank is the weight of water lost from the typical box itself due to being placed in a 50°C oven. This water loss is compensated for in the calculations for the weight percent of total product bleeding.

Box Weight After One Week	25.89	29.36	26.1	25.9	25.7
Difference in Box Weight	-0.71	2.82	-0.87	-0.84	-0.79
Weight Percent of composition bleeding <sup>2</sup>	-----	29.42%	0.013%	0.011%	0.006%

22. The results above demonstrate clearly that Example 1 of Voelkel which comprises approximately 20% GMS and approximately 40% of Dimethyldi(oleoylisopropyl)ammonium methosulfate in a softener-to-substrate ratio of about 86:35, produced a substantial level of box bleed. Specifically, about 29.42% by weight of the product bled onto the box.

23. There is a substantial difference in the amount of box bleed between the compositions described in the cited Voelkel reference (29.42%) and those of the instant patent application (0.013%). Indeed, no or negligible box bleed was observed in the Box Bleed Test using the compositions of the instant invention. This difference is significant, and in my opinion, quite unexpected.

24. I consider the nearly complete lack of box bleed produced by the compositions of the invention to be significant and completely unexpected in view of the results produced by the Voelkel reference, Example 1 (as suggested by the Examiner for comparison during the Interview of March 4, 2004).

25. I hereby declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like are

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<sup>2</sup> Weight Percent Of Product Bleeding; Calculated by:

(Difference in box weight for the example - difference in box weight for the blank)  
Total Weight of Active impregnated on Dryer Sheets (approximately 12 grams)

made punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 6-15-04

Matthew Levinson, Ph.D.

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